Indian J. Phys. 54B, 378–383 (1980)

Laser probe technique and its use for domain structure measurement in alanine doped triglycine sulphate

M M Pradhan and R K Garg

National Physical Laboratory, New Delhi-110012

Abstract: Focussed laser beam has been used to probe domain structure in pyroelectric crystals. A three mm dia He-Ne laser beam is converged to 50 μm size by a lens system and scanning is carried out on the surface plane of the pyroelectric crystals. Local heating produced in a small area by laser beam, causes pyroelectric signal which is processed in a phase sensitive lock-in amplifier and recorded. Phase of the signal reverses as the laser spot moves over the antiparallel domains. Depending upon the domain, pyroelectric signal varies from positive to negative side of the zero line in the record. Successive line scans are recorded which reveal the domain structure of the crystal plate Pyroelectric signal is taken by depositing electrodes on the crystal plate, and connecting them to a F.E.T. preamplifier circuit. The front electrode is made of semitransparent gold costing and the back electrode has thick gold coating

Laser probe technique may also be used to reveal 90° domain walls such as found in barium titanate Pyroelectric signal becomes null when the laser spot moves over 90° domain region during scanning.

Using laser probe technique, domain structure of L-Alanine doped triglycine sulphate (LATGS) has been studied. It is concluded that the value of coercive field in LATGS is greater than in TGS.

1. Introduction

Different types of techniques are used to reveal the domain structure of pyroelectric crystals. Crystal surface is cleaned and chemically etched. Pattern of domains is then viewed by spreading colloidal suspension (Pearson and Feldmann 1959), polarising microscope (Merz 1952, 1954; Little 1955) or multiple beam interference (Bhide *et al* 1961, 1963, 1965). Recently scanning electron microscope (Noriyuki 1973), second harmonic generation (Dolino 1973) and nematic liquid crystals (Furuhata 1973) have also been used.

For many practical applications such as pyroelectric infrared detector, pyroelectric vidicon or pyroelectric analysers, crystal plates are coated with metallic films. None of the above mentioned techniques may then be used to obtain domain pattern. For such a case Hadni and coworkers (Hadni *et al* 1972, 1974, 1975) used pyroelectric map method where antiparallel domains are obtained on intensity modulated C.R.O. display. The purpose of this paper is to describe the use of focussed laser beam and lock-in amplifier to obtain structure of anti-parallel domains. The method can as well be used to reveal 90° domains.

2. Laser probe

Modulated laser beam is used to produce thermal shocks on a pyroelectric crystal plate. A low wattage He-Ne laser beam is converged by a lens system to a small size of 50 microns from an original diameter of 3 mm. The beam is modulated at a low frequency of 11 Hz by a mechanical chopper and its intensity may be reduced to a desired level with the help of neutral density filter.

Crystal plates are cut perpendicular to polar axis, and polished. Various thicknesses ranging from 50 μm to 5 mm have been used for such measurements. Plates are coated under vacuum with semitransparent layer of gold on one side and thick layer of gold on the other side. Radiation is made to fall on the semi-transparent side. It allows to heat the crystal plate, as well as forms an electrode to carry the charges developed by alteration in spontaneous polarisation. The two electrodes of the crystal plate are shunted by a load resistance and then connected to a F.E.T. preamplifier circuit. Schematic arrangement of the system is shown in Figure 1.

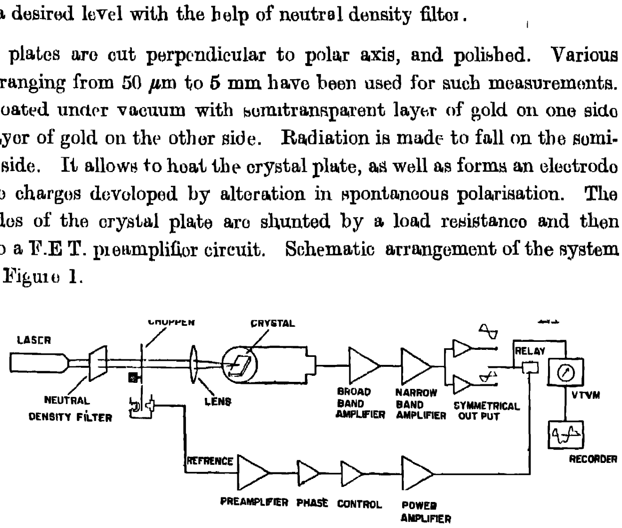


Figure 1. Schematic diagram of pyroelectric signal measurement by Laser probe technique

3. Scanning system

The crystal plate along with its preamplifier circuit is mounted on a X-Y shifter. Movement along X-axis is done by a synchronous motor which drives the crystal at 4 μm/sec. Movement along Y-axis is done manually with an accuracy of one micron. After complition of one line scan, the crystal is moved in Y direction for another line scan. The laser beam falls on the crystal plate along Z-axis.

4. Reference signal

To operate the lock-in amplifier, a reference signal is obtained with the help of a lamp and photo-diode through the mechenical chopper which modulates the laser beam. The signal is amplified and passed through delay circuit for phase control. Finally it is used to operate the vibrating lead of a relay. This vibrating load of the relay rectifies the pyroelectric signal to provide a d.c. output for recording purpose.

5. Signal processing

Pyroelectric signal which is developed in the crystal plate by the modulated laser beam, is amplified by a tuned amplifier and divided into two symmetrical outputs. The vibrating reed of the relay alternately touches the two outputs and takes the information for half cycle on each. Initially the relay is set by phase control in such a way that when it touches one of the two outputs, the signal in it is going from zero to positive side. At the end of half cycle, the vibrating reed comes to the other symmetrical output. At this instant, here also the signal is going from zero to positive side. Thus the vibrating reed of the relay only sees positive halves of the signal and an average positive rectified output is obtained. However, within the same setting, if the phase of the incoming signal changes by 180°, the average rectified output will be negative. The average output signal will be zero if the phase of the incoming signal changes by 90°.

The rectified output from the relay is given to a VTVM where signal is averaged over a certain time period and finally it is recorded on a strip chart recorder.

Different line scans along X-axis are obtained by displacing the crystal in a step of 50 μm after each scan. Domain pattern of the crystal is obtained by combining these line scans.

6. The pyroelectric signal

The focussed laser beam provides local heating of the crystal surface in a small region. Since the beam is modulated, the crystal is subjected to alternate heating depending upon the exposure and nonexposure of the crystal to radiation. The heat hemisphere expands inside the crystal plate and the rate of expansion depends upon the thermal conductivity of the crystal. This results in a temperature gradient along the polar axis.

Pyroelectric materials have macroscopic electric polarisation even in the absence of an external electric field. The polarisation results from the alignment of internal microscopic electric dipoles in the material. This spontaneous polarisation is a function of the temperature and is proportional to the energy absorbed on the crystal.

Since the two electrodes of the crystal plate are connected through external circuit, free charges come to these electrodes to balance the polarisation charge.

Thus they generate a current in the circuit. The pyroelectric current i_p is given by the rate of change of polarisation $(\partial p_s/\partial t)$ with time :

$$i_p = A\left(\frac{\partial p_s}{\partial t}\right)$$

$$= A\left(\frac{\partial p_s}{\partial T}\right)_E\left(\frac{\partial T}{\partial t}\right) = Ap\left(\frac{\partial T}{\partial t}\right) \tag{1}$$

where A is the electrode area and T is the instantaneous temperature. The pyroelectric coefficient $p = \left(\frac{\partial P_s}{\partial T}\right)_E$ is the rate of change of polarisation with temperature at constant stress.

In case of crystal with antiparallel domains, the spontaneous polarisation P_s is the vector sum of the spontaneous polarisation (P_{s+}) contributed by the positive domain and spontaneous polarisation (P_{s-}) contributed by the negative domain. When this crystal is scanned by the laser beam and pyroelectric signal is analysed by lock-in amplifier as described above, the pyroelectric signal varies from positive to negative side or vice versa of the zero line depending upon the nature of the domain over which the laser spot is falling. When the laser spot passes from one domain to other, the intensity of the pyroelectric signal becomes zero at the time of intermediate position of the laser spot.

Crystal having 90° domains walls also may be studied by this technique. Here in addition to positive and negative pyroelectric signal, one obtains the zero signal when the laser spot scans the 90° domain.

7. Domain pattern of LATGS

Laser probe technique was used to study domain pattern of *L*-Alanine doped triglycine sulphate (LATGS) crystals. Crystals of LATGS were grown from solution containing 25% *L*-Alanine and 75% Triglycine Sulphate. We have used 100 μm thick crystal plate which was cut perpendicular to polar axis. Pyroelectric signal obtained by scanning the crystal plate was analysed to obtain the domain pattern. Figure 2(a) shows a line scan of LATGS. For comparison purpose, a typical line scan of pyroelectric signal of a triglycine sulphate (TGS) crystal plate is shown in Figure 2(b). Domain pattern of LATGS crystal plate was obtained by joining different line scans taken in succession and without considering the intensity of the signal. The positive and negative domains are represented by black and white patches in Figure 3.

It is observed that the intensity of pyroelectric signal within the region of a domain, is quite constant and the domain size in LATGS is larger than that of TGS. Intensity of pyroelectric signal was found to vary in side the domain in

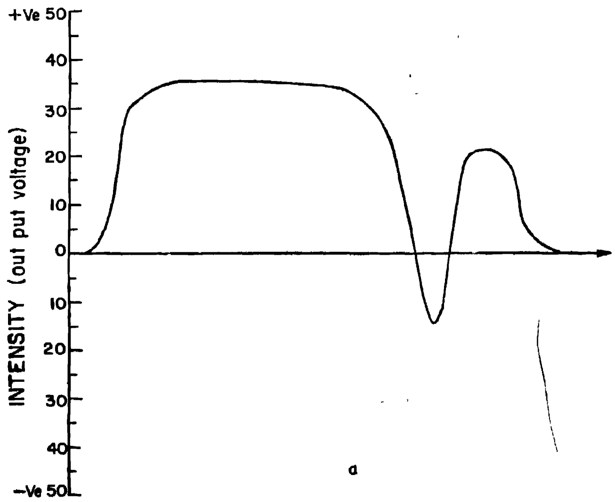


Figure 2(a). Line Scan of pyroelectric signal of LATGS

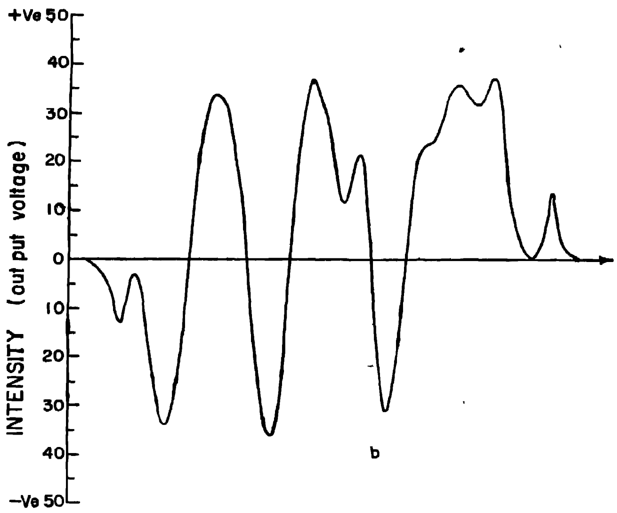
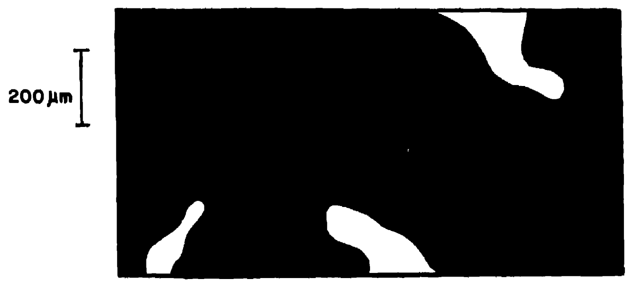


Figure 2(b). Line Scan of pyroelectric signal of TGS.



200 µm

Figure 3. Domain pattern of LATGS crystal plate.

TGS crystal (Pradhan and Garg 1977). It is related to the existance of reversible domains (Hadni *et al* 1973), in crystals of TGS. Nowhere the existence of such reversible domains was observed in LATGS. Lock (Lock 1971) has studied various properties of LATGS. He has shown that LATGS is always in a fully poled state in the absence of large external field. It is therefore concluded that the electric field created by thermal hemisphere of the laser spot was ineffective to reverse the domain because of large coercive field of the crystal.

Acknowledgment

We are grateful to Dr. V. G. Bhide for inspiring interest in this work.

References

Bhide V G and Bapat N J 1961 *Physica* **27** 631
——— 1963 *J. Appl. Phys.* **34** 81
Bhide V G and Chimulgund N D 1965 *Ind. J. Pure. Appl. Phys.*, **3** 253
Dlino G 1973 *Appl. Phys. Letters*, **22** 123
Furuhatay and Toriyama K 1973 *Appl. Phys. Letters*, **23** 361
Handi A and Thomas R 1974 *Optics Commun.*, **10** 366
Handi A and Thomas R 1972 *Ferroelectrics*, **4** 39
Hadni A, Lambert J P Pradhan M M and Thomas R 1973 *Infrared Phys*, **13** 225
Hadni A and Thomas R 1975 *Physica Status Solidi (a)* **31** 71
Little E A 1955 *Phys. Rev.*, **98** 978
Lock P J 1971 *Appl. Phys. Letters*, **19** 390
Merz W J 1952 *Phys. Rev.* **88** 421
Merz W J 1954 *Phys. Rev.* **95** 690
Noriyuki X and Nakatani X 1973 *Jap. J. Appl. Phys.*, **12** 313
Pearson G L and Feldmann W L 1959 *J. Phys. Chem. Solids*, **9** 28
Pradhan M M and Garg R K 1977 *Optics Commun*, **20** 422